



Docket No: 218880PCT

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

IN RE APPLICATION OF: : GROUP ART UNIT: 1755

Kunihiko TASHINO, et al. :

SERIAL NO: 10/030,120 : EXAMINER: J. Brown

FILED: January 24, 2002 :

FOR: SOLID CATALYST COMPONENT  
AND CATALYST FOR  
POLYMERIZATION OF OLEFINS

**LETTER SUBMITTING EXECUTED DECLARATION**

COMMISSIONER FOR PATENTS  
ALEXANDRIA, VIRGINIA 22313

SIR:

Application(s) respectfully submit herewith the original executed Declaration under 37 C.F.R. §1.132 to replace the unexecuted Declaration under 37 C.F.R. §1.132, which was originally filed in the U.S. Patent and Trademark Office on June 3, 2004.

Respectfully submitted,

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Appln. No. 10/030,120  
Declaration, 37 CFR 1.132



DOCKET NO: 218358US0PCT

IN THE UNITED STATES PATENT & TRADEMARK OFFICE

IN RE APPLICATION OF :  
Kunihiko TASHINO, et al : EXAMINER: BROWN, J.  
SERIAL NO: 10/030,120 :  
FILED: JANUARY 24, 2002 : GROUP ART UNIT: 1755  
FOR: SOLID CATALYST COMPONENT :  
AND CATALYST FOR  
POLYMERIZATION OF OLEFINS

DECLARATION 37 CFR 1.132

COMMISSIONER FOR PATENTS  
P. O. BOX 1450  
ALEXANDRIA, VIRGINIA 22313-1450

SIR:

Now comes Runihiko Tashino who deposes and says that:

- 1) I am one of the inventors of the above-identified application.
- 2) I have been employed by Toho Titanium Ltd. where I have been engaged in the development of magnesium-titanium Ziegler-Natta catalysts.
- 3) Presently, I am employed by the Toho Catalyst Co., Ltd., which is a subsidiary of Toho Titanium co., Ltd.
- 4) I have read the outstanding Office Action from the U.S. Patent Office and each of the references cited therein against the present claims.

5) That in order to demonstrate the superior property aspects of olefin polymerization catalysts prepared from the solid catalyst material of the present invention over catalysts prepared as described in the cited and applied references, the following comparative evidence has been obtained.

#### Experiment 1

The same experiment as described in Example 4 of the present specification was conducted, except that 0.13 g of 3-ethylcatechol was used as the alcohol component instead of 0.11 g of catechol. The content of titanium in the resulting solid catalyst component was 2.8 wt %. The results of polymerization are shown in the following Table.

#### Experiment 2

The same experiment as described in Example 4 of the present specification was conducted, except that 0.16 g of 3,5-diethylcatechol was used as the alcohol component instead of 0.11g of catechol. The content of titanium in the resulting solid catalyst component was 3.0 wt %. The results of polymerization are shown in the following Table.

#### Experiment 3

The same experiment as described in Example 4 of the present specification was conducted, except that 0.15 g of 3-chlorocatechol was used as the alcohol component instead of 0.11g of catechol. The content of titanium in the resulting solid catalyst component was 3.1 wt %. The results of polymerization are shown in the following Table.

#### Experiment 4

The same experiment as described in Example 4 of the present specification was conducted, except that 0.17 g of 3-bromocatechol was used as the alcohol component instead of 0.11g of catechol. The content of titanium in the resulting solid catalyst component was 3.3 wt %. The results of polymerization are shown in the following Table.

#### Experiment 5

The same experiment as described in Example 9 of this present specification was conducted, except that 0.18 g of 1,2,3-naphthalenetriol was used as the alcohol component instead of 0.16 g of 2,3-naphthalenediol. The content of titanium in the resulting solid catalyst component was 3.0 wt %. The results of polymerization are shown in the following Table.

#### Experiment 6

The same experiment as described in Example 3 of the present application was conducted, except that 0.11 g of thiophenol was used as the alcohol component instead of 0.1 g of 1,2-benzene dithiol. The content of titanium in the resulting solid catalyst component was 3.5 wt %. The results of polymerization are shown in the following Table.

#### Experiment 7

The same experiment as described in Example 3 of the present specification was conducted, except that 0.14 g of 3-methyl thiophenol was used as the alcohol component instead of 0.14 g of 1,2-benzene dithiol. The content of titanium in the resulting solid catalyst component was 3.7 wt %. The results of polymerization are shown in the following Table.

#### Experiment 8

The same experiment as described in Example 3 of the present specification was conducted, except that 0.15 g of 3-ethyl thiophenol was used as the alcohol component instead of 0.14 g of 1,2-benzene dithiol. The content of titanium in the resulting solid catalyst component was 3.7 wt %. The results of polymerization are shown in the following Table.

#### Experiment 9

The same experiment as described in Example 3 of the present specification was conducted, except that 0.16 g of 3-methyl benzene-1,2-dithiol was used as the alcohol component instead of 0.14 g of 1,2-benzene dithiol. The content of titanium in the resulting solid catalyst component was 3.8 wt %. The results of polymerization are shown in the following Table.

#### Experiment 10

The same experiment as described in Example 3 of the present specification was conducted, except that 0.17 g of 3-ethyl benzene-1,2-dithiol was used as the alcohol component instead of 0.14 g of 1,2-benzene dithiol. The content of titanium in the resulting solid catalyst component was 3.8 wt%. The results of polymerization are shown in the following Table.

#### Experiment 11

The same experiment as described in Example 3 of the present specification was conducted, except that 0.16 g of 3-chlorothiophenol was used as the alcohol component

instead of 0.14 g of 1,2-benzene dithiol. The content of titanium in the resulting solid catalyst component was 3.8 wt %. The results of polymerization are shown in the following Table.

#### Experiment 12

The same experiment as described in Example 3 of the present specification was conducted, except that 0.17 g of 3-chlorobenzene-1,2-dithiol was used as the alcohol component instead of 0.14 g of 1,2-benzene dithiol. The content of titanium in the resulting solid catalyst component was 3.5 wt %. The results of polymerization are shown in the following Table.

#### Experiment 13

The same experiment as described in Example 3 of the present specification was conducted, except that 0.18 g of 3-bromobenzene-1,2-dithiol was used as the alcohol component instead of 0.14 g of 1,2-benzene dithiol. The content of titanium in the resulting solid catalyst component was 3.4 wt %. The results of polymerization are shown in the following Table.

#### Comparative Experiment 1

The same experiment as described in Example 1 of the present specification was conducted, except that 0.11 g of o-cresol was used as the alcohol component instead of 0.094 g of phenol. The content of titanium in the resulting solid catalyst component was 3.0 wt %. The results of polymerization are shown in the following Table.

Comparative experiment 2

The same experiment as described in Example 3 of the present specification was conducted, except that 0.05 g of methyl mercaptan was used as the alcohol component instead of 0.14 g of 1,2-benzene dithiol. The content of titanium in the resulting solid catalyst component was 3.4 wt %. The results of polymerization are shown in the following Table.

Comparative experiment 3

A catalyst component was prepared by the same method as described in Example 1 of Murata (US Patent 4, 950, 630), except that 0.15 g of 3-chlorocatechol was used as the alcohol component instead of 2.0 ml of 2,2,2-trichloroethanol was conducted. The preparation of a polymerization catalyst and polymerization were conducted by the same procedures described in Example 4 of the present specification. The content of titanium in the resulting solid catalyst component was 3.4 wt %. The results of polymerization are shown in the following Table.

Comparative experiment 4

A catalyst component was prepared by the same method as described in Example 1 of Murata (US Patent 4,950,630), except that 0.11 g of thiophenol was used as the alcohol component instead of 2.0 ml of 2,2,2-trichloroethanol. The preparation of polymerization catalyst and polymerization were conducted by the same method as described in Example 3 of the present application. The content of titanium in the resulting solid catalyst component was 3.4 wt %. The results of polymerization are shown in the following Table.

The results of Examples 1 to 9 are also shown in Table.

	Polymerization activity (g-PP/g-cat.)	HI (wt%)	MI (g/100min)
Example 1	46,200	98.9	3.2
Example 2	51,100	98.5	7.9
Example 3	45,700	98.1	8.9
Example 4	52,100	98.7	6.5
Example 5	51,900	98.1	8.2
Example 6	50,600	98.2	9.5
Example 7	47,500	98.4	7.9
Example 8	52,100	98.1	7.3
Example 9	52,800	98.6	6.3
Experiment 1	51,500	98.5	6.8
Experiment 2	50,700	98.6	6.5
Experiment 3	55,600	98.4	8.1
Experiment 4	57,900	98.1	7.8
Experiment 5	52,100	98.5	6.9
Experiment 6	48,900	98.9	9.1
Experiment 7	50,200	98.4	8.5
Experiment 8	49,600	98.6	7.9
Experiment 9	49,800	98.7	8.6
Experiment 10	53,400	98.3	9.5
Experiment 11	52,100	98.2	11
Experiment 12	55,100	98.1	11
Experiment 12	56,800	98.0	10
Example 1	46,200	98.9	3.2
Comparative experiment 1	45,100	98.7	3.0
Comparative experiment 2	41,200	98.0	7.5
Comparative experiment 3	25,700	98.1	8.0
Comparative experiment 4	28,200	98.5	7.5

6) The data presented above with respect to Examples 2 and 4 to 9 of the specification and Experiments 1 to 5 of the declaration show that the polypropylene polymer products obtained as a result of using catalysts prepared from solid catalyst components prepared in turn from hydroxyl group-containing hydrocarbon compounds which have two or three hydroxyl groups and from a dialkoxo magnesium compound have remarkably higher



melt indexes than the melt index of a polymer obtained from a catalyst prepared from a solid catalyst component prepared in turn from a hydroxyl group-containing hydrocarbon compound which has one hydroxyl group and from a dialkoxy magnesium compound as described in Example 1 of the present specification.

The catalysts prepared from a solid catalyst component prepared in turn from a hydroxyl group-containing hydrocarbon compound and from a dialkoxy magnesium compound as described in Examples 2 and 4 to 9 of the specification and Experiments 1 to 5 of the declaration show remarkably higher activities than the catalyst prepared from a solid catalyst component prepared in turn from a hydroxyl group-containing hydrocarbon compound and magnesium metal as described in Comparative Experiment 3.

The catalysts prepared from a solid catalyst component prepared in turn from a mercapto group-containing hydrocarbon compound and a dialkoxymagnesium (present claim 2) described in Example 3 of the specification and Experiments 6 to 13 of the declaration show remarkably higher activities than the catalyst prepared from a solid catalyst component prepared in turn from a mercapto group-containing hydrocarbon compound and magnesium metal as described in Comparative Experiment 4.

7) The undersigned petitioner declare further that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of this application or any patent issuing thereon.

2004年 7月12日 9時29分

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8) Further, deponent saith not.

JUNE 25, 2004

Date

Kunihiko Tashino

Signature